Standard Operating Procedure for the Determination of Radium 226 in Drinking Water

1.0 Scope and Applicability

This method is used to determine the level of Radium 226 in drinking water. Increased levels over time in drinking water may be a cause of cancer. This method is nearly specific for Radium 226.

2.0 Summary of Method

The radium in the drinking water sample is concentrated and separated by co-precipitation with barium as the sulfate. The precipitate is dissolved in EDTA reagents, placed in a sealed bubbler, and stored for ingrowth of Radon-222. The scintillations are counted in an Alpha Scintillation counter. The half life of Ra 226 is 1602 years and the half of Rn 222 is 3.83 days.

3.0 Definitions

ppm	parts per million
mL	milliliter
EPA	Environmental Protection Agency
pCi	picoCurie

4.0 Interferences

The gaseous alpha-emitting radionuclides of radon 219 and radon 220 interfere and these are found in water contaminated by such industrial wastes as uranium mill elements.

5.0 Safety

Use safety precautions when transporting acid from other locations. Use the safety rules for radioactive materials. This method does not address all safety issues associated with its use. The laboratory and it's director are responsible for maintaining a safe work environment and current awareness of the file of OSHA regulations regarding the safe handling of the chemicals specified in this method. A reference file of material safety data sheets (MSDS's) should be available to all personnel involved in this analysis.

5.1 Contamination Control

5.1.1 Do not mouth pipette radioactive materials

5.1.2 Do not eat, drink, smoke, or apply cosmetics where radioactive materials are used

5.1.3 All radiological work should be done in a designated, posted area.

5.1.4 Thoroughly wash hands after handling radioactive materials.

5.2 Radioactive Spills Clean-Up

5.2.1 Spills should be quickly wiped up and the contaminated cleaning materials

placed in radioactive waste. This waste is disposed of using a commercial waste disposal company. Clean-up should be performed while wearing disposable protective covering prior to testing.

5.2.2 For containment, place absorbent material around the edges of the spill.

5.2.3 Blot up as much of the spill as possible, placing soaked materials in a waste bag.

5.2.4 Wash the area with dilute Contrad 70 solution. Wipe a small area with a dry filter paper and count by liquid scintillation.

5.2.5 The amount of removeable activity should be less than twice background.

6.0 Equipment and Supplies

Note: Brand names, suppliers, and part numbers are cited for illustrative purposes only. No endorsement is implied. Equivalent performance may be achieved using equipment and materials other than those specified here, but demonstration of equivalent performance that meets the requirements of this method is the responsibility of the laboratory.

6.1 Alpha Scintillation Counter.

6.2 Lucas Scintillation Cells.

6.3 Radon Emanation Apparatus.

6.4 Centrifuge.

6.5 40 mL glass centrifuge tubes

7.0 Reagents and Standards

7.1 15 N Ammonium Hydroxide (concentrated).

7.2 Ascarite drying reagent, 8-20 mesh.

7.3 Barium carrier, 16 mg/ml: Dissolve 2.846 gm $BaCl_2H_2O$ in distilled water, add 0.5 ml 16N HNO_3, and dilute to 100 ml with distilled water.

7.4 EDTA Reagent, 25 M: Dissolve 20 g NaOH in about 750 ml of distilled water, heat slowly, and add 93 g $Na_2C_{10}H_{14}O_8N_2$ 2 (H_2O) (disodium ethyl-enedinitrilo-acetate-dihydrate) while stirring. After the salt is in solution, filter through coarse filter paper and dilute to one liter.

7.5 Nitric Acid, 50%.

7.6 Helium Gas.

7.7 Glacial Acetic Acid.

7.8 Hydrochloric Acid, 12 N (concentrated).

7.9 Magnesium perchlorate, $Mg(ClO_4)_2$, reagent grade.

7.10 Sulfuric Acid, 18 N and 0.1 N.

7.11 Standard Radium-226 solution, approximately 50 pCi/ml.

8.0 Sample Collection, Preservation and Storage

The sample is acidified to a pH of 2 using nitric acid when the sample is collected. The sample's pH is checked with pH paper to verify the acidification and recorded in the sample pH log book.

9.0 Quality Control

9.1 Use 1000ul of spiking solution to spike a 1000ml sample, using pCi value of the spiking solution. Spike 10% of all samples.

9.2 Duplicate 10 % of all samples.

10.0 Calibration and Standardization

10.1 Calibration of alpha scintillation counters.

10.1.1 Count empty instruments for 990 or 1000 minutes for a background count. Record this in the Instrument QC Log. If the count is above 10 recount, and if still high turn HV down slightly and recount. Then perform step 10.1.2 to verify that sample will be counted accurately.

10.1.2 Count Randam alpha scintillation standard for 1 minute two to three times per analysis season. The cpm should be within the two standard deviation range. If not the HV can be adjusted.

11.0 Procedure

11.1 Add 20 ml. 12N HCL, add 2 ml. barrium carrier to 1 liter of sample.

11.2 Cautiously and with vigorous stirring, add 20 ml. 18N H_2SO_4. Let stand overnight.

11.3 Aspirate and discard supernatant.

11.4 Slurry the precipitate and transfer to a centrifuge tube. Centrifuge and discard supernatant. Wash twice with 0.1N H_2SO_4 and discard washes.

11.5 Add 20 ml. EDTA reagent, 4 drops 15N NH_4OH and heat in a boiling water bath until the precipitate dissolves.

11.6 Transfer the solution to a radon bubbler. Open both stopcocks and deemanate (purge) the solution by slowly passing helium gas through the bubbler for 20 minutes.

11.7 Close the stopcocks, record date, hour, and minutes. Store the solution for 4-8 days for ingrowth of Rn_{22}.

11.8 At the end of the storage period, fill the upper half of an absorption tube with magnesium perchlorate and the lower half with ascarite. Attach the tube to the radon bubbler, then attach the scintillation cell.

11.9 Turn on vacuum, open stopcock to scintillation cell and open stopcock to drying tube. Evacuate, turn off vacuum and check for leaks.

11.10 Open inlet on bubbler, open outlet stopcock on bubbler very slightly and allow bubbling to proceed so that 15-20 minutes are required for de-emanation. Record date, hour, and minutes.

11.11 Toward the end of de-emanation, when vacuum is no longer effective, gradually increase helium gas pressure. When the system is at atmospheric pressure, close all stopcocks, shut off helium gas, disconnect the bubbler, and record date, hour, minutes. This is the beginning of Radon-222 decay and ingrowth of Radon-222 daughters.

11.12 Store the scintillation cell for at least four hours to ensure equilibrium. Count using the scintillation counter. Record the date, hour, and minutes.

11.13 After each analysis flush and evacuate the cell ten times and store with helium at atmospheric pressure.

11.14 Fill the bubbler to within 1.5 mm of the neck constriction with water.

11.15 Clean all glassware with the Contrad 70 decontaminating solution. Rinse with distilled water.

12.0 Data Analysis and Calculations

12.1 Calculation of sample activity

Radium-226 activity in picocurie per liter = $\frac{\text{net cpm x C}}{\text{A x B x K x V}}$

Netcpm = cpm- background cpm.

C = Counting interval in minutes. (Counting time in minutes equals T_3)

$$C = \frac{\lambda t_3}{1-e^{-\lambda t_3}}$$

A = Decay time in minutes. (count start minus beginning de- emanation equals t_2).

B = ingrowth time in minutes. (End de-emanation minus purge equals T_1).

λ = .693/t 1/2 t 1/2 for Rn 222=5504.09 min.
λ = .693/5504.09= 0.0001259
V = Volume of sample
K = 2.2 X E (Converting dpm to pCi)

12.2 K Factor Calibration (Use approximately 50 pcCi of Radium 226 in bubbler)

$$E = \frac{C}{A(1-e^{\lambda t_1})(e^{-\lambda t_2})}$$

E = Calibration constant, includes de-emanation efficiency and counting efficiency of cell.

C = Net count rate in cpm.

A = Activity of Ra226 in bubbler in dpm.

t_1 = ingrowth time (end de-emanation minus purge)

t_2 = decay time (count start minus beginning de-emanation).

λ = .0001259 (1 pCi = 2.22 dpm)

13.0 Pollution Prevention
This method uses radioactive material which has to be disposed of through decay to

background or a disposal service.

14.0 Waste Managemant

The only waste is radioactive materials discussed above. For further information on waste management consult the Waste Management Manual for Laboratory Personnel and Less is Better: Laboratory Chemical Management for Waste Reduction, both available from the American Chemical Society's Department of Government Relations and Science Policy, 1155 16th Street N.W. , Washington D.C. ,20036

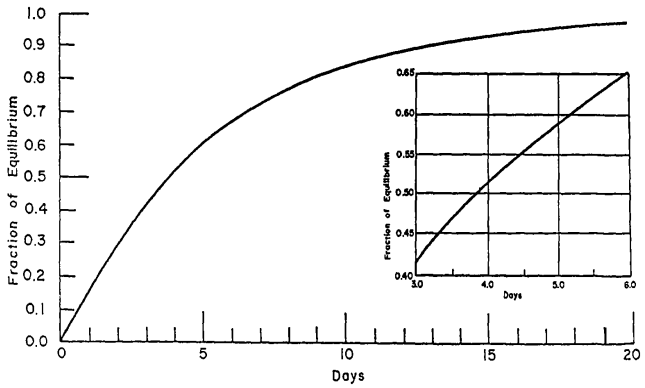
15.0 References

EPA-600/4-75-008 (Revised) March 1976

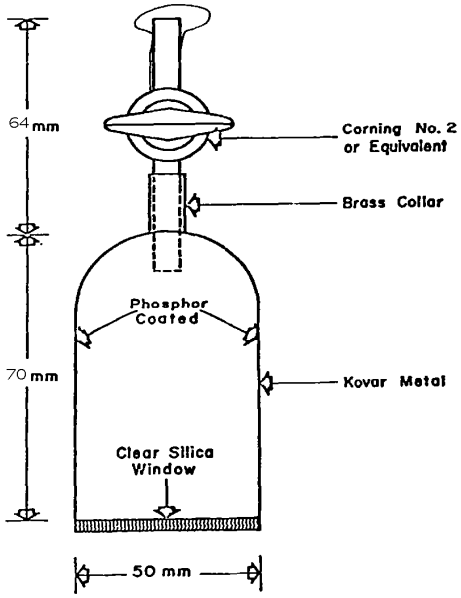
16.0 Tables, Diagrams, Flowcharts, Validation Data and Addtional Information Documentation Records

All information and values are recorded on a worklist, and raw data sheets are attached. The pH of each sample is recorded in the pH record book. Worklists are stored in a parameter log book in the bookcase in room 306.

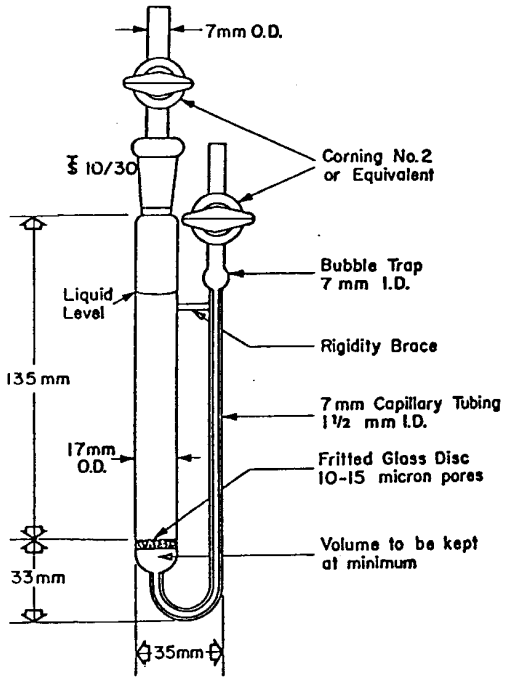
16.1 Radon Ingrowth Chart



16.2 Lucas Cell



16.3 Radon Bubbler



16.3 Deemanation Setup